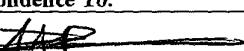


FORM PTO-1390 (REV 12-97)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		DATE: February 2, 2001 ATTORNEY'S DOCKET NUMBER: 54389USA8A.005 U.S. APPLICATION NO. 09/762109 (if known, see 37 C.F.R. 1.5)	
INTERNATIONAL APPLICATION NO. PCT/US99/18654	INTERNATIONAL FILING DATE 17 August 1999 (17.08.99)	PRIORITY DATE CLAIMED 17 August 1998 (17.08.98)	
TITLE OF INVENTION ELECTRODES			
APPLICANT(S) FOR DO/EO/US Shunsuke Takaki and Toshihiro Suwa			
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:			
1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. 2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. 3. <input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1). 4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19 th month from the earliest claimed priority date. 5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)). <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau). <input type="checkbox"/> has been transmitted by the International Bureau. <input checked="" type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US). 6. <input type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)). 7. <input checked="" type="checkbox"/> Amendment to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)). <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau). <input type="checkbox"/> have been transmitted by the International Bureau. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. <input checked="" type="checkbox"/> have not been made and will not be made. 8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). 9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)). 10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).			
Items 11. to 16. below concern other document(s) or information included:			
11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. 12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. 13. <input checked="" type="checkbox"/> A FIRST preliminary amendment. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment. 14. <input type="checkbox"/> A substitute specification. 15. <input type="checkbox"/> A change of power of attorney and/or address letter. 16. <input type="checkbox"/> Other items or information:			

Certificate of Express Mailing	
Under 37 CFR 1.10, I certify that this entry of a PCT application into US National Stage is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service addressed to the Commissioner for Patents, Washington, DC 20231, on the date indicated below.	
Express Mail Mailing Label No. EL 597089585US	Signature of Person Mailing Application <i>Colleen M. Wagner</i>
Date of Deposit February 2, 2001	Printed Name of Person Mailing Application Colleen M. Wagner

U.S. APPLICATION NO. (if known) see 37 C.F.R. 1.5 09/762109	INTERNATIONAL APPLICATION NO.:PCT/US99/18654	ATTORNEY'S DOCKET NUMBER 54389USA8A.005			
17. <input checked="" type="checkbox"/> The following fees are submitted: Basic National Fee (37 CFR 1.492(a)(1)-(5)):		Calculations PTO Use Only			
<input checked="" type="checkbox"/> Search Report has been prepared by the EPO or JPO \$860 <input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) .. \$690 <input type="checkbox"/> No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445 (a)(2)) \$710 <input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$1,000 <input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4) \$100					
ENTER APPROPRIATE BASIC FEE AMOUNT =		\$ 860.00			
Surcharge of \$ 130 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).		\$			
Claims	Number Filed	Number Extra	Rate		
Total Claims	18 -20 =	= 0	x \$ 18	\$	
Independent Claims	2 -3=	= 0	x \$ 80	\$	
MULTIPLE DEPENDENT CLAIM(S) (IF APPLICABLE)		x \$ 270	\$		
TOTAL OF ABOVE CALCULATION		\$ 860.00			
Reduction by $\frac{1}{2}$ for filing by small entity, if applicable. A Small Entity Statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28)		\$			
SUB TOTAL		\$			
Processing fee of \$ 130 for furnishing the English translation later the <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest priority date (37 CFR 1.492(f)).		\$			
TOTAL NATIONAL FEE		\$			
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property		\$			
TOTAL FEES ENCLOSED		\$			
		Amount to be refunded	\$		
		charged	\$		
a. <input type="checkbox"/> A check in the amount of \$ _____ to cover the above fees is enclosed.					
b. <input checked="" type="checkbox"/> Please charge my Deposit Account No. 13-3723 in the amount of \$ 860.00 to cover the above fees. A duplicate copy of this sheet is enclosed.					
c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 13-3723 A duplicate copy of this sheet is enclosed					
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.					

Send All Correspondence To:

Signature: 

Name: John A. Burtis	Registration Number: 39,924
Address: Office of Intellectual Property Counsel 3M Innovative Properties Company P.O. Box 33427 St. Paul, Minnesota 55133-3427	

Patent
Docket No.: 54389USA8A.005

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:
Shunsuke Takaki and Toshihiro Suwa

Serial No.: Unknown
Filed: Unknown
For: ELECTRODES

Group Art Unit:
Examiner:

PRELIMINARY AMENDMENT

Commissioner for Patents
Washington, D.C. 20231

Dear Sir:

Please amend the above-identified application as follows:

In the claims:

Please cancel claims 1-17 and substitute therefor the following claims:

18. An electrode adapted for attachment to an adherend comprising:
 - (a) an electrode support having a first and a second opposed surface,
 - (b) a conductor supported by the electrode support, and
 - (c) a conductive adhesive layer comprising a hydrophilic phase and a hydrophobic phase that is disposed upon a major portion of the first, opposed surface of the electrode support and the conductor,

wherein at least a portion of the conductive adhesive layer is hot-pressed to enhance the adhesion strength of the portion to an adherend.

19. An electrode according to claim 18, wherein the conductor is either in the form of a layer of conductive material disposed upon at least a portion of the first, opposed surface of the electrode support, or, the conductor is in the form a conductive stud that passes through a portion of the first, opposed surface of the electrode support and completely through the electrode support to project below the second, opposed surface of the electrode support.

20. An electrode according to claim 18, wherein the electrode further comprises a release liner disposed upon an exposed surface of the conductive adhesive layer.

21. The electrode according to claim 18, wherein the portion of the conductive adhesive layer that has been hot-pressed is a perimetrical portion.

22. The electrode according to claim 18, further comprising an indentation that separates the heat-pressed portion of the conductive adhesive layer from the remaining portion of the conductive adhesive layer.

23. An electrode according to claim 18, wherein the conductor is in the form of a layer of conductive material and the area of the conductor is essentially coextensive with the area of the electrode support.

24. An electrode according to claim 18, wherein the conductor comprises a conductive ink.

25. An electrode according to claim 18, wherein the conductive adhesive layer comprises: (a) a hydrophilic phase comprising hydrophilic polymer material, an electrolyte, and a humectant, and (b) a hydrophobic phase comprising hydrophobic polymer derived from the polymerization of hydrophobic monomer or oligomer in the presence of a surfactant and the hydrophilic phase.

26. An electrode according to claim 25, wherein the conductive adhesive layer consists essentially of: (a) a hydrophilic phase comprising hydrophilic polymer material, an electrolyte, and a humectant, and (b) a hydrophobic phase comprising hydrophobic polymer derived from the polymerization of hydrophobic monomer or oligomer in the presence of a surfactant and the hydrophilic phase.

27. An electrode according to claim 25, wherein the hydrophilic polymer material is selected from the group consisting of polymers containing one or more polyethylene glycol groups or polymers containing one or more pyrrolidone groups.

28. An electrode according to claim 25, wherein the electrolyte is selected from the group consisting of aqueous solutions of-. potassium chloride, sodium chloride or lithium chloride.

29. An electrode according to claim 25, wherein the humectant is selected from the group consisting of propylene glycol or sodium DL-pyrrolidonecarboxylate.

30. An electrode according to claim 25, wherein the hydrophobic polymer comprises interpolymerized units derived from one or more of the following monomers: acrylic acid, isoocetyl acrylate, 2-ethylhexyl acrylate and n-butyl acrylate.

31. An electrode according to claim 18, wherein the adherend is mammalian skin.

32. An electrode according to claim 25, wherein the adherend is mammalian skin.

33. A method of improving the adhesion strength of a conductive adhesive layer comprising a hydrophilic phase and a hydrophobic phase by hot-pressing a portion of the conductive adhesive layer.

34. The method according to claim 33, wherein the hot-pressed portion is a perimetrical portion.

35. The method according to claim 33, further comprising forming pinholes in the portion of the conductive adhesive layer to be hot-pressed prior to hot pressing.

It is believed that this Application is in condition for allowance and a notice to that effect is respectfully requested.

Respectfully submitted,

Registration Number	Telephone Number
39,924	(651) 736-4235
Date	
February 2, 2001	

Signature

Printed Name
John A. Burtis

Office of Intellectual Property Counsel
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ELECTRODES

Field of the Invention

The invention relates to electrodes, and particularly biomedical electrodes, having a conductive adhesive layer for attaching the electrode to an adherend (like mammalian skin), wherein a portion of the conductive adhesive layer is treated to provide improved adhesion to the adherend over the untreated portion of the conductive adhesive layer.

Background of the Invention

Heretofore, there has widely been used a biomedical electrode comprising an electrode supporting substrate, a stud-shaped, Ag/AgCl conductor, and a cohesive and non-adhesive conductive gel composition such as that disclosed in Unexamined Patent Publication (Kokai) No. 56-501108, WO81/00785, U.S. Patent No. 4,406,827. Since the conductive gel has no adhesion strength, a wide backing tape was required for fixing the electrode to the adherend.

Certain hydrophilic conductive adhesives as represented by Patent No. 2625179, Examined Patent Publication (Kokoku) No. 8-193494 and Patent Publication (Kokai) No. 7-501101 have been developed. These materials made it easier to apply and fix electrodes without using a backing tape for fixation. These conductive adhesives have adequate initial adhesion strength, but absorption of sweat from skin can reduce the adhesive's adhesion strength, and the edge portion of the electrode may delaminate from the skin. Adsorption of large quantities of sweat into the adhesive can convert the conductive adhesive into a gel state and the resulting adhesive may have essentially no adhesion strength. In such instances, the electrode easily moves on the skin which creates artifact noise, thereby obscuring readouts. Intense body movement of the subject, such as is sometimes the case when an electrocardiogram is taken, can create so much noise that a clear electrocardiogram can not be recorded.

Patent Publication (Kokai) No. 9-509196 describes a very useful conductive adhesive that has a hydrophilic phase and a hydrophobic phase and good physical properties. The hydrophilic phase of the adhesive contributes to the conductivity of the adhesive but it does not contribute as greatly to the adhesion strength of the adhesive as

does the hydrophobic phase. Accordingly, conductivity and adhesiveness must be balanced when formulating the adhesive for a particular electrode application. When conductivity of the adhesive must be very high, the hydrophilic phase may need to constitute a larger fraction of the adhesive, and this can adversely impact the long term adhesion strength of the adhesive. Accordingly, it would be useful to improve the adhesion strength of hydrophilic/hydrophobic phase adhesives to improve the long term wear properties of electrodes made using such adhesives without compromising electrode conductivity.

Brief Description of the Drawings

10 Fig. 1 is a cross-sectional view of one electrode embodiment of the invention that comprises a conductor in the form of a sheet or layer.

Fig. 2 is a cross-sectional view of another electrode embodiment of the invention that comprises a conductor in the form of a stud.

15 Fig. 3 is a perspective view showing one embodiment of a heat block that can be used to hot-press the conductive adhesive in the present invention.

Fig. 4 is a plan view of yet another electrode embodiment wherein the perimetrical portion of a conductive adhesive layer was hot-pressed using the heat block of Fig. 3 to provide a groove or depression between the hot-pressed portion of the conductive adhesive layer and the non-hot-pressed portion.

20

Summary of the Invention

In one aspect, the invention provides an electrode adapted for wear on an adherend, such as mammalian skin, comprising: (a) an electrode support having a first and a second opposed surface; (b) a conductor that is supported by the electrode support (for example, 25 by being disposed upon or through said first opposed surface); and (c) a conductive adhesive layer that is disposed upon a major portion (and preferably essentially all of the first, opposed surface of the electrode support and covering a major portion (and preferably essentially all) of the surface of the conductor. At least a portion of the conductive adhesive layer, and preferably a perimetrical portion, has been treated with heat and 30 pressure to improve the adhesion properties of the conductive adhesive to the adherend.

The conductive adhesive comprises, and preferably consists essentially of, a hydrophilic phase and a hydrophobic phase.

In preferred embodiments of the electrode, the electrode support supports a conductor either: (i) in the form of a layer of conductive material (e.g., conductive ink) disposed or coated upon at least a portion of the first opposed surface of the electrode support, or, (ii) in the form of a conductive stud that passes completely through a portion of the electrode support to project below the second opposing surface of the electrode support. The conductive adhesive layer is disposed upon a major portion of the first, opposed surface of the electrode support such that it covers at least a major portion (and preferably essentially all) of the surface of the conductor whether in the form of a layer, or a stud. In some embodiments, the stud may also project above the first opposing surface of the electrode support.

Preferably, a release liner is provided upon the surface of the conductive adhesive layer to protect the exposed surface of the adhesive layer until the electrode is ready to be applied to an adherend. Prior to applying the electrode to an adherend, the release liner is removed to expose the conductive adhesive layer.

In another aspect, the invention provides a method of improving the adhesion properties of a coating or layer of a conductive adhesive composition that comprises, and preferably consists essentially of, a hydrophobic and hydrophilic phase, by applying heat and pressure to a portion of said coating or layer.

Detailed Description of the Invention

Figure 1 shows one electrode embodiment of the invention. The electrode 10 comprises an electrode support 11 having two, opposed surfaces 12, 13, wherein a conductor 14, in the form of a sheet or layer of conductive material (e.g., a conductive ink coating), is disposed upon a portion of a first, opposed electrode support surface 12. Preferably, the area of the conductor 14 is coextensive with the area of the first, opposed electrode support surface 12. Provided on a portion of the exposed surface of the conductor 14, is a layer of a conductive adhesive 15. A release liner 16 is laminated to the exposed surface of the conductive adhesive layer 15. A portion of the conductor 14 that is not coated with conductive adhesive, together with an adjacent portion of the electrode

support 11, forms a tab 18 for attachment to a lead wire (not shown). A portion, and preferably a perimetrical portion, of the conductive adhesive layer 15 is hot-pressed according to the method described herein to improve its adhesion properties over the properties of the untreated portion of the conductive adhesive layer 15.

5 Figure 2 shows another electrode embodiment of the invention wherein the electrode 20 comprises an electrode support 21 with two, opposed surfaces 22, 23, and a conductor 24 in the form of a conductive stud. The conductor 24 passes through a portion of a first, opposed electrode support surface 22, and completely through the electrode support 21 to project below the second, opposed electrode support surface 23. A layer of 10 conductive adhesive is provided both over the first, opposed surface of the electrode support 22, and the surface of a conductor portion 26 that projects above the electrode support surface 22 to form a conductive adhesive layer 27. The surface of a second conductor portion 28 projecting below the second, opposed electrode support surface 23, serves as a connector for a lead wire (not shown). A portion, and preferably a perimetrical 15 portion, of the conductive adhesive layer 27 is hot-pressed according to the method described herein to improve the adhesion properties of the hot-pressed portion. Also shown is a release liner 29 disposed upon the exposed surface of the conductive adhesive layer 27.

20 In the heat-pressing process, heat can be applied directly to the conductive adhesive layer, or indirectly to the release liner. Typically, a heated block with a small area, such as that on a micro-press, is used to apply heat. An embodiment of a machined, metal block useful in heat pressing the conductive adhesive coating is shown in Figure 3. The block 30 has a rectangular, outer portion 31 and a cut-out 32. Disposed around the entire perimeter of the cut-out 32, is a raised projection 33 which can be of various shapes other than the 25 one depicted in Figure 3. The block 30 is heated to the desired temperature for example, by using a continuous or impulse heat source, ultrasonic waves, a radio frequency heat source, a lamp heater or the like. The temperature the block is heated will depend upon the formulation of the adhesive, and the thickness of the adhesive coating, but preferably, the block should be heated at a temperature from about 50 to 200°C, and preferably from 90 to 30 170°C. A temperature, which causes mobility of the surfactant in conductive adhesive, is preferred. The heated block is then applied to the conductive adhesive layer with a

pressure of about 0.5 to 4 kg/cm². The pressure should be selected so as not to break the conductive adhesive layer or to squeeze the adhesive layer off of the electrode. The pressing time should be selected based on the type of materials used in the construction of the electrode and the temperature of the heater. Typical pressing times are from about 0.1 to 10 seconds, and preferred times are about 1 to 5 seconds. When the pressing time is too long, the adhesion strength of the adhesive can be adversely affected.

Preferably, a heating block similar to that shown in Figure 3 is used to heat press the conductive adhesive layer. The block is positioned over the conductive adhesive layer such that the portion of the layer to be heat pressed is located under the outer portion 31, and the portion of the layer that is not to be heat pressed is located under the cut-out 32. An electrode having a structure similar to that depicted in Fig. 4 (shown without release layer) will result. The electrode 40 comprises an electrode support 41 with two, opposed surfaces 42, 43. Disposed on a first, opposed surface of the electrode support 42, is a sheet-like conductor 44 that is preferably coextensive with the area of the first, opposed electrode support surface 42. Disposed upon the conductor 44, is a conductive adhesive layer 45 that was heat pressed using the heat block shown in Fig. 3. The conductive adhesive layer has a heat-pressed perimetrical portion 46, which underlies the outer portion 31 of the heat block 30 during heat pressing. The conductive adhesive layer also has a groove or indentation 47 that is adjacent to the inner perimeter of the heat-pressed perimetrical portion 46, and the outer perimeter of a non-heat pressed portion of the conductive adhesive layer 48. The non-heat-pressed portion underlies the cut-out 32 of the heat block 30 during heat pressing. The groove 47 was formed by the projection 33 on the heat block 30. Also provided is a tab portion 49 that is not coated with conductive adhesive and is formed from the adjacent portions of the electrode support 41 and the conductor 44. The groove helps prevent the migration or diffusion of water from the non-heat pressed adhesive portion 48 into the heat pressed adhesive portion 46, thereby helping to maintain the improved adhesive properties obtained by heat pressing. Preferably, the volume of the groove or indentation should amount to less than about 10% of the volume of the non-heat pressed portion of the conductive adhesive layer. Preferably, such an indentation can be obtained by using a block with a convex protrusion 0.1 to 2 mm in height, and for an adhesive coating about 1 mm thick, a heat block protrusion of about 0.4

to 0.6 mm in height will form a clear groove between the hot pressed and non-heat pressed portions of the conductive adhesive layer. Alternatively, the formation of the groove and the heat pressing of the adhesive can be conducted in separate steps; although combining the steps as previously described is preferred.

5 If too great an area of the adhesive conductive layer is heat-pressed, the adhesion of the electrode to the skin can be too great and can cause pain when the electrode is removed from the skin. On the other hand, the heat-pressed adhesive area must be large enough to provide the desired adhesive wear properties for the electrode. Preferably, adhesion strength should be greater than 110 g (as measured in a 180 degree peel test using a sample width of 25.4 mm and a peel rate of 300 mm/min.) and less than an amount that will cause pain when the electrode is removed from the skin. Generally, 180 degree peel test values greater than about 500 g (measured according to the aforementioned test parameters) will result in painful electrode removal. Accordingly, 180 degree peel test values of about 110 to 500 g are preferred, and most preferably, values are about 150 to 500 g. Because heat 10 pressing can also reduce the water content of the conductive adhesive and impair the adhesive's conductivity, the heat-pressed area must be selected such that an acceptable overall conductivity for the conductive adhesive layer is maintained. Preferably, the absolute area of the heat-pressed portion of the conductive adhesive layer is at least about 1 cm². More preferably, the heat-pressed portion will be between about 1 to 4 cm². It is 15 preferred to heat press a perimetrical portion of the conductive adhesive layer because this increases the adhesive properties of the part of the electrode typically subjected to the most delaminating force (e.g., such as lead wire tension). Preferably, the width of the heat-pressed perimetrical portion for an electrode designed for short wearing periods will be about 1 to 3 mm. The heat-pressed, perimetrical portion width for electrodes designed for 20 long wear, or for situations in which the subject will move during wear (e.g., stress tests), is preferably about 5 to 20 mm.

Heat pressing may also be carried out by applying heat to the conductive adhesive layer in a heated pattern (e.g., lattice patterns, wave patterns, etc.).

30 Preferably, pinholes are introduced into the release liner prior to or during heat-pressing to permit the water vapor evolving during heat pressing to escape. The pinholes can be introduced prior to heat-pressing by piercing the release liner, and, if desired, the

underlying layers of the electrode, using a tool having a multiplicity of needle-like protrusions. The pierced area should approximately correspond to the area of the conductive adhesive layer that is to be heat-pressed. Alternatively, pinholes can be introduced during the heat-pressing process by using a heat block with a multiplicity of 5 needle protrusions located in the region of the block that will overly the portion of the conductive adhesive coating to be heat-pressed.

The electrode support can be selected from any flexible plastic film useful as a support for biomedical electrodes. Various commercially available plastic sheets about 10 μm to 1 mm thick may be used.

10 The conductor can be made of any of the conductive materials useful as conductors in biomedical electrodes. Various stud shaped and sheet-like conductors are known in the art and are suitable for use in the invention. Single or multilayer, sheet-like conductors may be used in the invention. One example of a useful conductor is a sheet coated with ink containing Ag and AgCl. An example of a commercially available sheet-like conductor that 15 is suitable for use in the invention is MSX-4510 manufactured by Minnesota Mining and Manufacturing Company, St. Paul, Minnesota, U.S.A. Other useful sheet-like conductors are described in PCT/US96/05938 and U.S. Patent No. 5,924,983 (which descriptions are herein incorporated by reference), where a two-layer conductive sheet comprising: (a) a low porosity base conductor layer made of a conductive coating composition containing a 20 hydrophobic polymer binder and carbon particles and, optionally, Ag particles, and (b) a porous top conductor layer, formed on the base conductor layer, that is made of a conductive coating composition containing a polymer binder, Ag/AgCl particles and carbon particles.

25 Any release liner, such as those obtained by coating a paper sheet or plastic film, that are useful in biomedical electrodes, can be used in this invention. Typically, such materials are plastic or paper sheets coated or impregnated with a material that imparts or enhances the release properties of the sheet.

30 The conductive adhesive layer comprises, and preferably consists essentially of, or consists of, a hydrophilic phase and a hydrophobic phase. Particularly preferred examples of such adhesives are disclosed in the Japanese Patent Publication (Kokai) No. 9-509196, WO95/20634, and U.S. Patent Nos. 5,409,966 and 5,438,988 (which descriptions are

incorporated by reference herein). A hydrophilic polymer material, an electrolyte, and a humectant for preventing evaporation of the electrolyte and water are generally contained in the hydrophilic phase. As the hydrophilic polymer material, a polyethylene glycol group-containing material having good ion conductivity, for example, methoxypolyethylene glycol or polymer derived from N-vinylpyrrolidone may be used. Such a hydrophilic polymer can be obtained by adding an ultraviolet polymerization initiator (e.g., 1-(4-(2-hydroxyethoxy)-phenyl)-2-hydroxy-2-methoxy-1-propane-1-one, 4-trimethylaminomethylbenzophenone hydrochloride, etc.) to a monomer or oligomer for the hydrophilic polymer material and performing the polymerization reaction. As the electrolyte, an aqueous solution of 5 potassium chloride, sodium chloride, lithium chloride or the like may be used. As the humectant, propylene glycol, sodium DL-pyrrolidonecarboxylate or the like can be used. A hydrophobic polymer material is contained in the hydrophobic phase. As the hydrophobic polymer, for example, there may be used copolymers of acrylic acid and isoctyl acrylate, 2-ethylhexyl acrylate, n-butyl acrylate and the like. Such a hydrophobic polymer can be 10 obtained by polymerizing the monomer or oligomer for the hydrophobic polymer, in a surfactant stabilized hydrophobic phase, that is contained in a hydrophilic phase. The polymerization can be carried out by exposing the polymerizable composition to UV 15 radiation. In such a case, an ultraviolet polymerization initiator for hydrophobic phase should be included in the polymerizable composition. As the above surfactant, there can be used polyoxyethylene oleyl ether, ammonium salt of α -sulfo- ω -(1-((nonylphenyl)methyl-2-(2-propenoxy)ethoxy-(polyoxy-1,2-ethanediyl)), ammonium salt of sulfated 20 polyoxyethylene alkyl ether and sodium salt of sulfosuccinic alkyl(C₈₋₂₀) alkenyl(C₃₋₄) ester. Examples of ultraviolet polymerization initiators for the hydrophobic phase include: 1-hydroxy-cyclohexyl-phenylketone and 2,2-dimethoxy-2-phenylacetophenone.

25 The conductive adhesive can be produced in the following manner. The aforementioned components of the hydrophilic phase and hydrophobic phase are mixed to prepare a liquid composition. This composition is purged with a nitrogen gas to drive out dissolved oxygen. A reinforcing material, such as a non-woven fabric, is placed on a silicone-coated surface of a transparent polyester liner, and, after impregnating it with or 30 applying the liquid to the reinforcing material at a thickness of about 0.2 to 2 mm, another layer of polyester liner is placed on the reinforcing material such that its silicone-coated

surface is brought into contact with the liquid. The composite is then exposed to UV radiation (e.g., 500 to 3000 mJ) to effect the desired level of cure in the adhesive. Subsequently, one of the polyester liner layers is peeled off and the conductive adhesive layer is laminated to the conductor and electrode support composite. The polyester liner that remains adhered to the other side of the conductive adhesive layer can be left adhered to the adhesive layer such that it becomes the release liner of the electrode. Thereafter, the electrode can be hot pressed. Before or after the hot pressing step, the edge of the electrode can be trimmed (where the conductor is in sheet form) to form a tab which serves as an attachment for a lead wire.

10 The following examples are offered to aid in the understanding of the present invention and are not to be construed as limiting the scope thereof. Unless otherwise indicated, all parts and percentages are by weight.

Examples 1 to 3 and Comparative Example C1

15 Preparation of Electrodes

A biomedical electrode was produced by forming a conductor layer and a conductive adhesive layer on a flexible plastic film support layer. The conductor layer was prepared according to the process described in U.S. Patent No. 5,924,983, which description is incorporated by reference herein. First, a 15 μm thick, sheet-like conductor having porosity (an adsorption surface area of nitrogen gas per unit area as measured by the BET method of $4.8 \text{ m}^2/\text{m}^2$) was formed on a transparent, 75 μm thick, polyester film (available from the Unitika Co. as "Emblet") by applying mixed ink for a base conductor layer and drying the coated film. The mixed ink for the base conductor layer was prepared by combining 98.5 parts of a first ink, 1 part of an Ag/AgCl ink available from Ercon Co. as "R-301"; and 0.5 parts of a curing agent available from Dow Chemical Co. as "PAPI 2027", using a mixer. The first ink which was prepared by combining 5.7 wt. percent of a conductive carbon having small porosity (BET value: $50 \text{ m}^2/\text{g}$) available from Mitsubishi Chemical Co. as "#3050B"; 2.5 wt. percent of a conductive carbon having large porosity (BET value: $980 \text{ m}^2/\text{g}$) available from Akzo Co. as "Ketchin Black EC"; 13.3 wt. percent of a urethane binder available from BF Goodrich Co. as "Estane 5703"; 47.5 wt. percent of a propylene glycol monomethyl ether acetate; 24.8 wt. percent of methyl ethyl ketone; and

6.2 wt. percent of toluene as a solvent. Then, an 8 μm thick, porous, top conductor layer (adsorption surface area of nitrogen gas per unit area measured by BET method: 24 m^2/m^2) was formed on the base layer only on the portion that would be in contact with the conductive adhesive by applying mixed ink for the top conductor layer and drying the 5 coating. The ink for the top conductor layer was prepared by combining: 84.5 parts of a second of ink, 15 parts of an Ag/AgCl ink available from Ercon Co. as R-301; and 0.5 parts of a curing agent available from the Dow Chemical Co. as "PAPI 2027", using a mixer. The second ink was prepared by combining: 5.2 wt. percent of conductive carbon having large porosity (BET value: 980 m^2/g available from Akzo Co. as "Ketchin Black EC"), 10 9.6 wt. percent of a urethane binder available from BF Goodrich Co. as "Estane 5703", 30.6 wt. percent propylene glycol monomethyl ether acetate, 43.7 wt. percent of methyl ethyl ketone, and 10.9 wt. percent of toluene as a solvent.

Drying of the base conductor layer was performed at the temperature between about 100 to 110°C and an air flow of 9 m/sec, and drying of the top conductor layer was 15 performed at temperatures between about 150 to 160°C and an air flow of 1.5 m/sec. Consequently, a conductor sheet comprising a polyester film, a base conductor layer with small porosity and relatively small Ag/AgCl content, and a top conductor layer with large porosity and relatively large Ag/AgCl content, was obtained.

On the conductor sheet, a conductive adhesive coating consisting of a hydrophilic 20 phase and a hydrophobic phase was formed by mixing raw materials in the proportion of the following table and performing the polymerization reaction.

Table 1

		Raw Materials	Example 1 (wt. %)	Example 2 (wt. %)	Example 3 (wt. %)
Hydrophobic Phase	Ultraviolet polymerization initiator	Irg. 184	0.05	0.07	0.06
	Monomer for hydrophobic polymer material	AA	13.92	15.43	13.75
		IOA	13.92	22.13	19.72
	Surfactant	Brij 97	17.9	--	--
		SE-10N	--	16.1	
		JS-2*	--	--	46.6
Hydrophilic Phase	Monomer for hydrophilic polymer material	MPEG-550MA	13.92	2.68	5.5
		AM90G	--	2.68	--
	Humectant	PPG	9.95	--	--
		PCA-Na	--	6.71	6
	Electrolyte	4% KCl	29.84	33.53	7.77
	Ultraviolet polymerization initiator	Irg. 2959	0.5	0.67	0.6
		Total	100	100	100

* Water content of JS-2 is 62 wt. %.

Acrylic acid (AA) and isoctyl acrylate (IOA) monomers and surfactant were combined, followed by polymerization using an ultraviolet polymerization initiator. As the surfactant, a polyoxyethylene oleyl ether ("Brij-97") available from Sigma Co. U.S.A. as "Brij-97", was used in Example 1; an ammonium salt of α -sulfo- ω -((nonylphenyl)methyl-2-(2-propenyloxy) ethoxy-(polyoxy-1,2-ethanediyl)) ("SE-10N"), available from Asahi Denka Chem Co. as "Adekreasorp SE-10N", was used in Example 2; and a sodium salt of sulfosuccinic alkyl (C_8 - 30) alkenyl (C_3 - 4) ester ("JS-2"), available from Sanyo Kasei Kogyo Co. as "Eleminol", was used in Example 3. 1-hydroxy-cyclohexyl-phenylketone ("Irg. 184"), available from Ciba Geigy Co. as "Irgacure 184", was used as the ultraviolet polymerization initiator for hydrophobic phase. The monomer methoxy polyethylene glycol 550 monoacrylate ("MPEG-550MA"), available from Satorn Chem. USA, was used to prepare the hydrophilic polymer material. In Example 2, an additional monomer, methoxy polyethylene glycol monoacrylate "AM-90G",

available from Shinnakamura Chem. Co. as "AM-90G", was also used to prepare the hydrophilic polymer material.

As the humectant, either propylene glycol ("PPG"), or sodium DL-pyrrolidonecarboxylate solution ("PCA-Na"), available from Ajinomoto Co. as "PCA-Na", were used. As the ultraviolet polymerization initiator for hydrophilic phase, 1-(4-(2-hydroxyethoxy)-phenyl)-2-hydroxy-2-methoxy-1-propane-1-one ("Irg. 2959"), available from Ciba Geigy Co. as "Irgacure 2959", was used.

A liquid mixture containing the above-described materials for the hydrophobic and hydrophilic phases was prepared. The resulting liquid was purged with a nitrogen gas to drive out dissolved oxygen, and a nonwoven fabric (available from Kimberly Clark Co. as "KC Tissue") was placed on a silicone-coated surface of a transparent polyester liner. After impregnating the fabric with the liquid to a thickness of 1 mm, another layer of the polyester liner was placed on top of the liquid coating, such that its silicone surface is brought into contact with the liquid. The resulting composite was cured by exposing it to UV radiation (about 2400 mJ/cm^2) using a low-pressure mercury lamp. Consequently, a 1 mm thick conductive adhesive layer consisting of a hydrophilic phase and a hydrophobic phase was obtained.

Evaluation of Adhesive Conductivity

Subsequently, the polyester liner sheets were peeled from the conductive adhesive layer, and the conductive adhesive layer was laminated to the conductor such that the top conductor layer contacted the conductive adhesive. Thereafter, the composite was cut to provide a conductive adhesive area of 5.5 cm^2 and a tab portion area of 1.6 cm^2 . Identical electrodes were laminated together and tested using the electrode characteristic test of the AAMI Society, U.S.A. The results of the conductivity tests are provided in Table 2 along with conductivity test results (Comparative Example C1) for a conductor layer/adhesive layer composite of the same construction except that a conductor adhesive commercially available from Minnesota Mining and Manufacturing Company, St. Paul, MN as RD-40 was used instead of a conductive adhesive with a hydrophobic phase and a hydrophilic phase. The data shows that the electrodes according to the invention have sufficient conductivity to be useful as biomedical electrodes.

Table 2

Test Measurements	AAMI specification	Example 1	Example 2	Example 3	Comparative Example C1
DC offset voltage	$\leq 100 \text{ mV}$	0.2 mV	-0.1 mV	-0.1 mV	-0.5 mV
AC impedance	$\leq 2000 \Omega$	500 Ω	499 Ω	400 Ω	422 Ω
Offset potential after 5 seconds since the completion of defibrillation	$\leq 100 \text{ mV}$	2.2 mV	9.3 mV	14.3 mV	24.1 mV
Recovery rate after 5 seconds since the completion of defibrillation	0 to -1.0 mV/s	-0.3 mV/s	-0.3 mV/s	-0.5 mV/s	-0.6 mV/s

Evaluation of Adhesive Peel Strength

Table 3: 180 degree peel test

Example No.	Peel strength (g/inch)
1	115
2	182
3	210
C1	247

The biomedical electrodes produced as described above were hot-pressed in the following manner. In each of the electrodes, pinholes having a pore diameter of 0.5 mm were formed through the entire surface of the release liner. The holes were spaced about 3 mm apart. The whole surface of the adhesive was hot-pressed by applying a hot press to the release liner at various temperatures under a pressure of 1 kg/cm^2 for 5 seconds. Each electrode was applied to the lower arm of a human body and pressed on to the skin. The 180 degree peel strength values were immediately measured by peeling the electrode from the skin at a rate of 300 mm/min. The results are provided in Table 4.

Table 4

Temperature (°C)	Peel Strength			
	Example 1 (g/in)	Example 2 (g/in)	Example 3 (g/in.)	C1 (g/in.)
Untreated	115	182	210	247
43	148	190	237	238
65	193	280	275	233
93	298	410	485	230
98	358	380	545	222
129	520	380	428	210
165	480	250	373	188
171	137	220	288	160

The data in Table 4 shows that the peel strength of the conductive adhesive increases as the hot press temperature increases up to a maximum hot press temperature.

5 The optimal hot pressing temperature appears to depend upon the surfactant used, but many effective temperatures are found out within the range from 90 to 170°C.

Furthermore, hot press temperatures greater than 170°C are generally not preferred because high temperatures can deteriorate the other components of the electrode (e.g., release liner or electrode support). An increase in adhesion strength was not achieved by 10 hot pressing the adhesive R-40D (see C1).

Electrodes identical to Example 1, Example 2 and Comparative Example C1 electrodes were prepared as described above, except that the electrode width was 22 mm, the area of the conductive adhesive was 5.5 cm², the area of the tab was 1.6 cm², and the thickness of the conductive adhesive was about 1 mm. Thereafter, some of the electrodes 15 were hot-pressed on the release liner side at 125°C and under a pressure of 2 kg/cm² for 1.4 seconds using a heat block with a protrusion 0.6 mm high for forming a groove. The protrusion formed grooves in the adhesive layer 1 mm wide, that separated perimetrical, hot-pressed adhesive portions from central, untreated, adhesive portions. The perimetrical, hot-pressed portions were about 1.5 mm wide. After pressing, the thickness of the 20 perimetrical, hot-pressed adhesive portions were about 0.5 mm. Two, identical, non-hot-pressed electrodes and two, identical, hot-pressed electrodes, respectively, were laminated together and the electrode characteristic test of AAMI Society, U.S.A. was performed on each pair of laminated electrodes. Although the effective area of conductive adhesive layer

of the hot-pressed electrode was reduced to about 61.8% of that of the untreated electrodes, no deterioration of electrical characteristics were measured. One of the non-hot-pressed electrodes, one of the hot-pressed electrodes and an electrode of identical construction except that RD-301 conductive adhesive was used instead of the hydrophilic/hydrophobic phase adhesive were applied to the lower arm of a human body and pressed by a 2 kg. Roller. The 180 degree peel strength for each electrode was immediately measured using a peel rate of 300 mm/min. The results are provided in Table 5. The data shows that the peel strength of electrodes prepared with the hydrophilic phase/hydrophobic phase adhesive improved when the adhesive was heat-pressed.

10

Table 5

Ex. No.	No Hot Press Peel Strength (g/in)	Hot Press Peel Strength (g/in)
1	138	162
2	230	313
C1	317	247

Example 4

An electrode was prepared by laminating a 1 mm thick conductive adhesive coating (identical to Example 1) to a conductor on a plastic film, and laminating a matted polyethylene tape (commercially available from Minnesota Mining and Manufacturing Co. U.S.A. as "Blenderm") to the plastic film surface of the tab portion on the opposite side of the conductor layer, and cutting the resulting laminate so that the electrode width was 34 mm, the area of the conductive adhesive was 13.6 cm^2 and the area of the tab portion was 1.44 cm^2 . Then, the tab portion was removed, and 0.5 mm diameter pinholes were formed in the laminate by piercing through the release layer in the 3 mm wide portion of the laminate to be hot pressed.

Thereafter, a heat block with a 0.6 mm high protrusion height for forming a groove was used to hot press the laminate and form a groove. In the transverse direction of the electrode, the width of the hot-pressed portion of the adhesive coating was 5 mm. In the longitudinal direction, the width of the hot pressed portion was 8 mm. The thickness of the peripheral, hot pressed portion of the adhesive layer was 0.4 mm. The width of the groove

in the adhesive layer was 1 mm. The hot pressing was performed by pressing either the electrode support or the release liner at a pressure of 2 kg/cm^2 for 1.4 seconds under several temperature conditions. Two, non-hot-pressed electrodes and two, hot-pressed electrodes, respectively, were laminated together and an electrode characteristic test of 5 AAMI Society, U.S.A. was performed on each laminated pair. Although the effective area of the hot-pressed conductive adhesive was reduced to about 32.4% of that of the untreated electrode, no deterioration of electrical characteristics was measured. The non-hot-pressed electrode and hot-pressed electrode were each applied to the lower arm of a human body and pressed by a 2 kg. roller. The 180 degree peel strength values for each 10 electrode were immediately measured using a peel rate of 300 mm/min. The results are provided in Table 6.

Table 6

Surface to be hot-pressed	180° Peel Strength (g/34 mm width)					
	No Heat Press	81°C Heat Press Temp	96°C Heat Press Temp	110°C Heat Press Temp	125°C Heat Press Temp	140°C Heat Press Temp
Electrode Support Layer	118	130	140	155	190	140
Release Liner	118	150	165	185	215	150

15 Regardless of the surface that is hot-pressed, the adhesion strength improved by hot pressing up to about 125°C, and good adhesion improvement can be obtained by applying the hot press to the release liner surface.

20 To examine the effect of pinholes, a pinhole-treated electrode and non-pinhole-treated electrode were hot-pressed on the release liner surface at temperatures of 110°C and 125°C. The skin adhesion strength of the electrode was measured in the same manner. The results are shown in Table 7. Good results were obtained when the pinhole treatment was performed.

Table 7

	180° Peel Strength (g/34 mm)		
	No Heat Press	110°C Heat Press	125°C Heat Press
No pinhole, treatment	118	110	185
Pinhole treatment	118	160	215

To examine the effect of the hot press pressure and the effect of the height of the protrusion during formation of the groove, the adhesion strength of the electrode obtained by hot-pressing the pinhole-treated electrode from the release liner surface using a heat block at the temperature of 125°C using various pressures and protrusion height was measured in the same manner. The degree of "squeeze-out" of the adhesive from the edge of the electrode after the heat press treatment is shown in Tables 8, 9 and 10.

10

Table 8

	Heat Press Pressure (kg)					
	No Heat Press	0.5	1.0	1.5	2.0	3.0
Peel strength (g/width of electrode: 34 mm)	118	150	220	190	185	--
Squeeze-out amount of adhesive	None	None	None	Small	Small	Large

Protrusion height: 0 ram
Hot pressing: 1.4 seconds

15

Table 9

	Heat Press Pressure (kg)					
	No Heat Press	0.5	1.0	1.5	2.0	3
Peel strength (g/width of electrode: 34 mm)	118	--	160	170	222	--
Squeeze-out amount of adhesive	None	--	None	None	None	Large

Protrusion height: 0.4 mm
Hot pressing: 1.4 seconds

Table 10

	Heat Press Pressure (kg)					
	No Heat Press	0.5	1.0	1.5	2.0	3
Peel strength (g/width of electrode: 34 mm)	118	--	--	185	215	--
Squeeze-out amount of adhesive	None	--	--	None	None	Large

Protrusion height: 0.6 mm

Hot pressing: 1.4 seconds

5 When a heat block having no protrusion for forming a groove was used, the adhesion strength was improved up to 1 kg/cm². When the pressure was higher than 1 kg/cm², the adhesive was broken and squeezed out from the periphery of the electrode. When a heat block having a protrusion was used, the adhesion strength was improved at heat press pressures up to 2 kg/cm². However, when the pressure was increased to 10 3 kg/cm², a large amount of the adhesive was squeezed out from the periphery of the electrode.

15 A hot-pressed electrode and an identical electrode made without hot pressing (both made using the adhesive of Example 1), were applied to the lower arm of a human body and pressed by a 2 kg. Then a lead wire was attached to the tab portion of each electrode and a weight of 100 g was suspended from the lead wire. The subject operated an ergometer (bicycle for stress test) under a load of 100 W. The time until the electrodes peeled off was measured. The average peeling time for the hot-pressed electrodes was 12.5 minutes while the average peeling time of the electrodes prepared without hot pressing was 10.5 minutes.

19-10-2000

PCT/US99/18654

MINNESOTA MINING AND MANUFACTURING CO.

Our Ref.: E 1206 PCT

US 009918654
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SIEBERTSTR. 4
81675 MÜNCHEN

Claims:

19. Okt. 2000

1. An electrode adapted for attachment to an adherend comprising:
 - (a) an electrode support having a first and a second opposed surface,
 - 5 (b) a conductor supported by the electrode support, and
 - (c) a conductive adhesive layer comprising a hydrophilic phase and a hydrophobic phase that is disposed upon a major portion of the first, opposed surface of the electrode support and the conductor, wherein a portion of the conductive adhesive layer is hot-pressed to enhance the adhesion strength of the
 - 10 portion to an adherend, and wherein a portion of the conductive adhesive layer is not hot-pressed.
2. An electrode according to claim 1, wherein the conductor is either in the form of a layer of conductive material disposed upon at least a portion of the first, opposed surface of the electrode support, or, the conductor is in the form a conductive stud that passes through a portion of the first, opposed surface of the electrode support and completely through the electrode support to project below the second, opposed surface of the electrode support.
- 20 3. An electrode according to any preceding claim, wherein the electrode further comprises a release liner disposed upon an exposed surface of the conductive adhesive layer.
4. The electrode according to any preceding claim, wherein the portion of the conductive adhesive layer that has been hot-pressed is a perimetrical portion.
- 25 5. The electrode according to any of the preceding claims, further comprising an indentation that separates the heat-pressed portion of the conductive adhesive layer from the portion of the conductive adhesive layer that
- 30 has not been hot-pressed.

6. An electrode according to any preceding claim, wherein the conductor is in the form of a layer of conductive material and the area of the conductor is essentially coextensive with the area of the electrode support.

5 7. An electrode according to any preceding claim, wherein the conductor comprises a conductive ink.

8. An electrode according to any preceding claim, wherein the conductive adhesive layer comprises:

10 (a) a hydrophilic phase comprising hydrophilic polymer material, an electrolyte, and a humectant, and
(b) a hydrophobic phase comprising hydrophobic polymer derived from the polymerization of hydrophobic monomer or oligomer in the presence of a surfactant and the hydrophilic phase.

15 9. An electrode according to claim 8, wherein the conductive adhesive layer consists essentially of:
(a) a hydrophilic phase comprising hydrophilic polymer material, an electrolyte, and a humectant, and
(b) a hydrophobic phase comprising hydrophobic polymer derived from the polymerization of hydrophobic monomer or oligomer in the presence of a surfactant and the hydrophilic phase.

20 10. An electrode according to any of claims 8 to 9, wherein the hydrophilic polymer material is selected from the group consisting of polymers containing one or more polyethylene glycol groups or polymers containing one or more pyrrolidone groups.

25 11. An electrode according to any of claims 8 to 10, wherein the electrolyte is selected from the group consisting of aqueous solutions of potassium chloride, sodium chloride or lithium chloride.

12. An electrode according to any of claims 8 to 11, wherein the humectant is selected from the group consisting of propylene glycol or sodium DL-pyrrolidonecarboxylate.

5

13. An electrode according to any of claims 8 to 12, wherein the hydrophobic polymer comprises interpolymerized units derived from one or more of the following monomers: acrylic acid, isoctyl acrylate, 2-ethylhexyl acrylate and n-butyl acrylate.

10

14. An electrode according to any of the preceding claims wherein the adherend is mammalian skin.

15. A method of improving the adhesion strength of a portion of a conductive adhesive layer comprising a hydrophilic phase and a hydrophobic phase by hot-pressing a portion of the conductive adhesive layer and not hot-pressing a portion of the conductive adhesive layer.

16. The method according to claim 15, wherein the hot-pressed portion is a perimetrical portion.

17. The method according to any of claims 15 to 16, further comprising forming pinholes in the portion of the conductive adhesive layer to be hot-pressed prior to hot pressing.

25

1/2

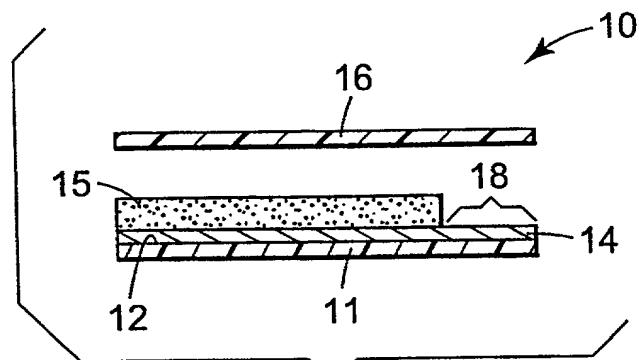


Fig. 1

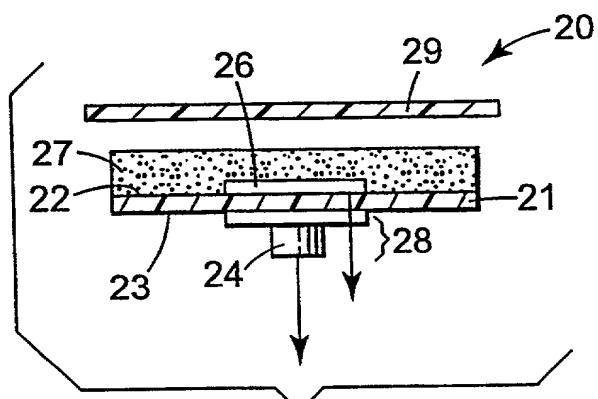


Fig. 2

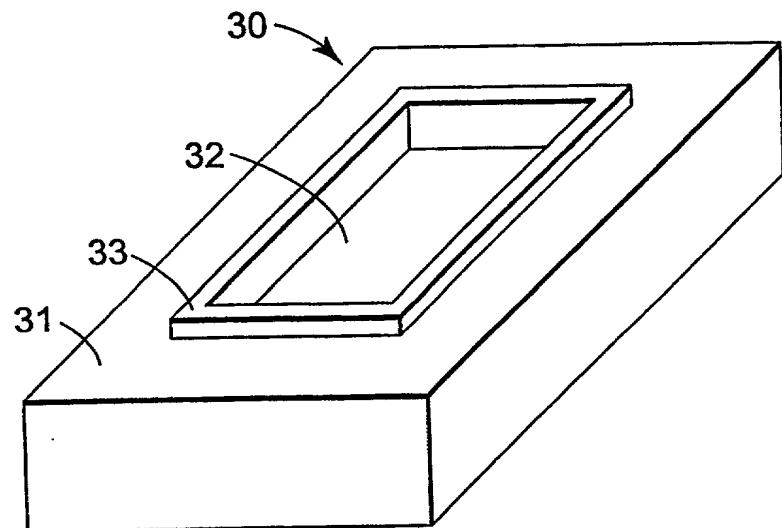


Fig. 3

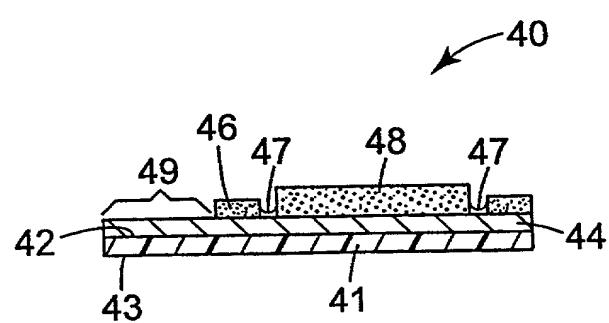
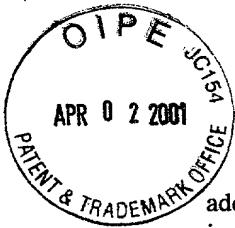


Fig. 4

**DECLARATION, POWER OF ATTORNEY, AND PETITION**

I, a below named inventor, depose and say that: (1) my residence, citizenship, and mailing address are indicated below; (2) I have reviewed and understand the contents of my patent application, including the claims, as amended by any amendment specifically referred to herein, which is identified as U.S. Patent Application Serial No. 09/762,109, filed February 2, 2001; and PCT International Patent Application Serial No. PCT/US99/18654 filed August 17, 1999, bearing Attorney Docket Number 54389PCT6A.003; (3) I believe that the other below named inventors and I are the original, first, and joint inventors or discoverers of the invention or discovery in

ELECTRODES

described and claimed therein and for which a patent is sought; and (4) I hereby acknowledge my duty to disclose to the Patent and Trademark Office all information known to me to be material to the patentability as defined in Title 37, Code of Federal Regulations, §1.56*, and that no application for patent or inventor's certificate on this invention or discovery has been filed by us or our legal representatives or assigns in any country foreign to the United States of America except Japan Application No. 10-230594 filed August 17, 1998 upon which we hereby claim foreign priority benefits under Title 35, United States Code Section 119.

I hereby appoint Gregory D. Allen (Reg. No. 35,048), Alan Ball (Reg. No. 42,286), Scott A. Bardell (Reg. No. 39,594), Carolyn A. Bates (Reg. No. 27,853), Dale A. Bjorkman (Reg. No. 33,084), Colene E. H. Blank (Reg. No. 41,056), Jennie G. Boeder (Reg. No. 28,952), William J. Bond (Reg. No. 32,400), Stephen W. Buckingham (Reg. No. 30,035), John A. Burtis (Reg. No. 39,924), Paul W. Busse (Reg. No. 32,403), Gerald F. Chernivec (Reg. No. 26,537), James D. Christoff (Reg. No. 31,492), Philip Y. Dahl (Reg. No. 36,115), Janice L. Dowdall (Reg. No. 31,201), Lisa M. Fagan (Reg. No. 40,601), Carolyn A. Fischer (Reg. No. 39,091), Yen T. Florczak (Reg. No. 45,163), Darla P. Fonseca (Reg. No. 31,783), John A. Fortkort (Reg. No. 38,454), Melanie G. Gover (Reg. No. 41,793), Christopher D. Gram, (Reg. No. 43,643), Gary L. Griswold (Reg. No. 25,396), Doreen S. L. Gwin (Reg. No. 35,580), Michael A. Hakamaki (Reg. No. 40,011), Karl G. Hanson (Reg. No. 32,900), Néstor F. Ho (Reg. No. 39,460), Rudolph P. Hofmann, Jr. (Reg. No. 38,187), Jeffrey J. Hohenshell (Reg. No. 34,109), Robert W. Hoke (Reg. No. 29,226), MarySusan Howard (Reg. No. 38,729), Stephen C. Jensen (Reg. No. 35,207), Robert H. Jordan (Reg. No. 31,973), Harold C. Knecht III (Reg. No. 35,576), Kent S. Kokko (Reg. No. 33,931), Douglas B. Little (Reg. No. 28,439), Eloise J. Maki (Reg. No. 33,418), Lisa M. McGeehan (Reg. No. 41,185), Matthew B. McNutt (Reg. No. 39,766), Michelle M. Michel (Reg. No. 33,968), William D. Miller (Reg. No. 37,988), Peter L. Olson (Reg. No. 35,308), Daniel R. Pastirik (Reg. No. 33,025), David B. Patchett (Reg. No. 39,326), Robert J. Pechman (Reg. No. 45,002), Carolyn V. Peters (Reg. No. 33,271), Scott R. Pribnow (Reg. No. 43,869), Ted K. Ringsred (Reg. No. 35,658), Steven E. Skolnick (Reg. No. 33,789), Robert W. Sprague (Reg. No. 30,497), Brian E. Szymanski (Reg. No. 39,523), James J. Trussell (Reg. No. 37,251), Lucy C. Weiss (Reg. No. 32,834), and Kimberly S. Zillig (Reg. No. 46,346) my attorneys and/or agents with full powers (including the powers of appointment, substitution, and revocation) to prosecute this application and any division, continuation, continuation-in-part, reexamination, or reissue thereof, and to transact all business in the Patent and Trademark Office connected therewith; the mailing address and the telephone number of the above-mentioned attorneys and/or agents are

Attention: John A. Burtis
Office of Intellectual Property Counsel
3M Innovative Properties Company
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St. Paul, Minnesota 55133-3427
Telephone No. (651) 736-4235

The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Wherefore, I pray for grant of Letters Patent for the invention or discovery described and claimed in the aforementioned specification and we hereby subscribe our names to the foregoing specification and claims, declaration, power of attorney, and this petition, on the day set forth below.

Shunsuke Takaki

March 20, 2001

Date

Shunsuke Takaki

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Toshihiro Suwa

March 21, 2001

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§1.56 Duty to disclose information material to patentability.

(a) A patent by its very nature is affected with a public interest. The public interest is best served, and the most effective patent examination occurs when, at the time an application is being examined, the Office is aware of and evaluates the teachings of all information material to patentability. Each individual associated with the filing and prosecution of a patent application has a duty of candor and good faith in dealing with the Office, which includes a duty to disclose to the Office all information known to that individual to be material to patentability as defined in this section. The duty to disclose information exists with respect to each pending claim until the claim is cancelled or withdrawn from consideration, or the application becomes abandoned. Information material to the patentability of a claim that is cancelled or withdrawn from consideration need not be submitted if the information is not material to the patentability of any claim remaining under consideration in the application. There is no duty to submit information which is not material to the patentability of any existing claim. The duty to disclose all information known to be material to patentability is deemed to be satisfied if all information known to be material to patentability of any claim issued in a patent was cited by the Office or submitted to the Office in the manner prescribed by §§ 1.97(b)-(d) and 1.98. However, no patent will be granted on an application in connection with which fraud on the Office was practiced or attempted or the duty of disclosure was violated through bad faith or intentional misconduct. The Office encourages applicants to carefully examine:

- (1) prior art cited in search reports of a foreign patent office in a counterpart application, and
- (2) the closest information over which individuals associated with the filing or prosecution of a patent application believe any pending claim patentably defines, to make sure that any material information contained therein is disclosed to the Office.

(b) Under this section, information is material to patentability when it is not cumulative to information already of record or being made of record in the application, and

- (1) It establishes, by itself or in combination with other information, a prima facie case of unpatentability of a claim; or
- (2) It refutes, or is inconsistent with, a position the applicant takes in:
 - (i) Opposing an argument of unpatentability relied on by the Office, or
 - (ii) Asserting an argument of patentability.

A prima facie case of unpatentability is established when the information compels a conclusion that a claim is unpatentable under the preponderance of evidence, burden-of-proof standard, giving each term in the claim its broadest reasonable construction consistent with the specification, and before any consideration is given to evidence which may be submitted in an attempt to establish a contrary conclusion of patentability.

(c) Individuals associated with the filing or prosecution of a patent application within the meaning of this section are:

- (1) Each inventor named in the application;
- (2) Each attorney or agent who prepares or prosecutes the application; and
- (3) Every other person who is substantively involved in the preparation or prosecution of the application and who is associated with the inventor, with the assignee or with anyone to whom there is an obligation to assign the application.

(d) Individuals other than the attorney, agent or inventor may comply with this section by disclosing information to the attorney, agent, or inventor.